

### **REMARKS**

The Official Action dated September 2, 2010 has been carefully considered.

Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claim 5 is amended to recite that the process uses reactants consisting essentially of alkali metal hydrosulfide, alkali metal hydroxide, and dihalo-aromatic compound, in organic amide solvent, in accordance with the teachings throughout the present application, and to include limitations from claim 19. It is believed that these changes are fully supported by the original specification, whereby entry of the present Amendment is in order and is respectfully requested.

In the Official Action, claims 5, 7, 10-13, and 17-19 were rejected under 35 U.S.C. 103(a) as being unpatentable over Miyahara et al, US 5,840,830. Although the Examiner asserted the claims were anticipated by Miyahara et al (page 5, penultimate line), it is believed that under 35 U.S.C. §103(a), the Examiner is asserting that Miyahara et al render the claims obvious. The Examiner asserted that Miyahara et al teach a process for producing a poly(arylene sulfide) (PAS) by polymerizing a sulfur source and a dihalo-aromatic compound in an organic amide solvent. The Examiner asserted that Miyahara et al teach many of the limitations of claim 5 and, while the Examiner acknowledged differences between the Miyahara et al process and the process of claim 5, the Examiner asserted that such differences did not prevent the finding of a prima facie case of obviousness. The Examiner relied on the Edmunds, Jr. et al. U.S. Patent No. 4,324,886 and the Campbell U.S. Patent No. 3,867,356 as teaching that it is known to employ alkali metal alkylaminoalkanoates as starting materials in the field of PPS preparation, and, specifically, that each of these references teaches the reaction of an organic amide such as NMP

with sodium hydroxide to form sodium N-methyl-4-aminobutyrate and reaction of this compound with sodium bisulfide (also known as sodium hydrosulfide).

This rejection is traversed and reconsideration is respectfully requested. Applicants submit that Miyahara et al do not teach or suggest, or make otherwise apparent to one of ordinary skill in the art, important limitations of the process of claim 5 which in combination provide PAS with improved properties, and that neither Edmunds, Jr. et al nor Campbell resolve the deficiencies of Miyahara et al and, in fact, these references demonstrate the unpredictability of the presently claimed process.

More particularly, by carefully controlling the reactants and reaction conditions, the present process allows the production in a single reactor of a desirable and versatile PAS from relatively inexpensive reactants, providing an economically viable process. The process for producing a PAS as defined in claim 5 employs reactants consisting essentially of alkali metal hydrosulfide, alkali metal hydroxide, and dihalo-aromatic compound, in organic amide solvent, and comprises four important process steps which are conducted in a single reactor and which in combination, provide a PAS having a desirable combination of improved properties as also set forth in claim 5.

Specifically, in the dehydration step (1), an organic amide solvent, and an alkali metal hydrosulfide and an alkali metal hydroxide, both as aqueous mixtures, are heated and reacted in a proportion of 0.95 to 1.02 mol of alkali metal hydroxide per mol of the alkali metal hydrosulfide, wherein distillate containing water is discharged to the exterior of the system and hydrogen sulfide formed upon the dehydration is discharged as a gas to the exterior of the system. As the discharge of hydrogen sulfide to the exterior of the system is directly linked with a weight loss of

sulfur amount in the system, it is evident that the discharged hydrogen sulfide is not recycled into the reaction system.

In the subsequent charging step (2), the total number of mols of (i) alkali metal hydroxide formed with hydrogen sulfide upon the dehydration, (ii) the alkali metal hydroxide added prior to the dehydration, and (iii) the alkali metal hydroxide added after the dehydration is 1.015 to 1.050 mols per mol of the charged sulfur source and the mols of water are controlled to provide 0.5 to 2.0 mols of water per mol of the charged sulfur source. The amount of the charged sulfur source is calculated out in accordance with the equation: [Charged sulfur source] = [Total moles of sulfur charged] – [Moles of sulfur volatilized out after dehydration], again evidencing that the discharged hydrogen sulfide is not recycled into the reaction system. Additionally, the polymerization is conducted in the defined first-stage polymerization step (3) and the defined second-stage polymerization step (4) to provide a polymer as claimed, in the same reactor in which the dehydration step is conducted.

The combination of the recited reactants and the defined steps (1)-(4) as set forth in claim 5 is important in providing a PAS polymer product in high yield and having good melt viscosity, low yellowness index, low dimer byproduct, good reactivity with aminosilane coupling agents, and low volatility from the alkali metal hydrosulfide and alkali metal hydroxide reactants in an economically viable process. The present process differs from Miyahara et al in several respects. While each difference alone may appear to the Examiner as minor, these differences in combination are significant and, along with the other limitations of claim 5, unpredictably provide a relatively low cost process and a high quality PPS product that are neither taught, suggested nor apparent from the teachings of Miyahara et al.

As noted in Applicants' previous response, it is preferred to use the claimed alkali metal hydrosulfide and alkali metal hydroxide as starting reactants as they are less expensive than the traditionally employed alkali metal sulfide. However, in the past, it has been difficult to produce a polymer having a good combination of properties in a stable manner using these reactants, as described in the present specification, beginning at page 2, line 17. Applicants discovered, as further described in the present specification, for example beginning at page 17, line 6, the alkali metal hydroxide unexpectedly reacts with the organic amide solvent in the dehydration heat treatment to form an alkali metal alkylaminoalkanoate, and the alkali metal hydrosulfide exists in the system in the form of a complex with the alkali metal alkylaminoalkanoate. While a part of the alkali metal hydrosulfide reacts with water to form hydrogen sulfide and an alkali metal hydroxide, the resulting hydrogen sulfide is discharged outside the system. The steps recited in claim 5 control these various reactions arising from the use of the alkali metal hydrosulfide and alkali metal hydroxide reactants to provide a stable reaction and a polymer product having a consistently good combination of properties as recited.

Miyahara et al disclose and exemplify a process for producing poly(arylene sulfide) wherein an alkali metal sulfide such as sodium sulfide is reacted with a dihaloaromatic compound, and wherein hydrogen sulfide vaporized off during the dehydration step is recovered and reused. Miyahara et al indicate that the sodium sulfide can be produced by in situ reaction of sodium hydrosulfide and sodium hydroxide (column 4, lines 61-64) and that these reactants can be used in equimolar amounts (column 6, lines 49-53). Importantly, Miyahara et al fail to exemplify any process wherein sodium hydroxide and sodium hydrosulfide are employed as reactants as required by present claim 5 or wherein sodium sulfide is formed in situ.

The Examiner has relied on Edmunds, Jr. et al and Campbell as teaching that it is known to employ alkali metal alkylaminoalkanoates as starting materials in the field of PPS preparation, and, specifically, that each of these references teaches the reaction of an organic amide such as NMP with sodium hydroxide to form sodium N-methyl-4-aminobutyrate and reaction of this compound with sodium bisulfide (also known as sodium hydrosulfide). However, neither Edmunds, Jr. et al nor Campbell teach a process wherein reactants consisting essentially of alkali metal hydrosulfide, alkali metal hydroxide, and dihalo-aromatic compound, in organic amide solvent, are employed, or a reaction sequence as presently claimed.

That is, Edmunds, Jr. et al disclose contacting an alkali metal hydroxide, lactam and water in the presence of an alkali metal carboxylate to prepare an alkali metal alkylaminoalkanoate, which is then reacted with a polyhalosubstituted aromatic compound, an organic amide solvent and hydrogen sulfide ( $H_2S$ ). Thus, the Edmunds, Jr. et al reaction sequence requires an alkali metal carboxylate and  $H_2S$  as reactants, neither of which are employed as reactants in the process of claim 5, and requires a separate step for preparation of the alkali metal alkylaminoalkanoate, without contact with an alkali metal hydrosulfide. Thus, Edmunds, Jr. et al teach different reactants and different reactions than used or suggested by Miyahara or as required by claim 5.

Similarly, Campbell discloses the reaction of a polyhalosubstituted aromatic compound, an alkali metal bisulfide, an alkali metal alkylaminoalkanoate and organic amide. Like Edmunds, Jr. et al, Campbell fails to disclose a reaction which employs sodium hydroxide and sodium hydrosulfide only with the polyhalo-aromatic compound and organic amide solvent as reactants, without a preformed alkali metal alkylaminoalkanoate. Thus, Campbell, like Edmunds, Jr. et al, teaches different reactants and different reactions than used or suggested by

Miyahara et al or as required by claim 5. In fact, these references actually teach away from the presently claimed process as neither Edmunds, Jr. et al nor Campbell teach, suggest or recognize that inexpensive reactants, namely alkali metal hydroxide and alkali metal hydrosulfide, can simply be dehydrated by heating in an organic amide solvent and then reacted with dihalo-aromatic compound to provide a PAS having an extremely advantageous combination of properties. To the contrary, Edmunds, Jr. et al and Campbell teach a separate reaction step and the use of additional reactants, and, consequently, the simplicity of the presently claimed process in providing the defined PAS is unpredictable. Accordingly, contrary to the Examiner's assertion in the Official Action that a person having ordinary skill in the art would have expected improved melt viscosities and yields of PAS produced when using NaOH and NaSH as starting materials, neither Edmunds, Jr. et al nor Campbell teach the use of NaOH and NaSH together as starting materials and therefore provide no expectation of improved melt viscosities and yields of PAS, or any other improvement, by use of equimolar amounts of NaOH and NaSH in the Miyahara et al process.

Applicants' previous response also noted, importantly, if the process of Example 1a of Miyahara et al uses an alkali metal hydrosulfide (NaSH) and an alkali metal hydroxide (NaOH) in equimolar amounts in place of the exemplified sodium sulfide in the dehydration reaction, the proportion of alkali metal hydroxide to alkali metal hydrosulfide is 1.032, and therefore not in the range of 0.95 to 1.02 mol as required by claim 5. The Examiner questioned this statement and the underlying calculation on the basis that the additional small amount of sodium hydroxide which is added in the working examples of Miyahara et al is added **after** the dehydration step. Applicants submit however that the calculations and statement are correct, as the additional small amount of sodium hydroxide which is added in the working examples of Miyahara et al after the

dehydration step is only used in the calculation of the total amount of NaOH for the NaOH/sulfur source ratio calculation. The only sodium hydroxide which is employed in Applicants' calculations for the sodium hydroxide/sodium hydrosulfide ratio is the sodium hydroxide which is initially charged and that which is added for the dehydration step as described at column 12, lines 16-21 of Miyahara et al:

"Operation was conducted in the same manner as in Comparative Example 1a except that this reaction apparatus was used, **30 g of 97% NaOH** were added to the 20-liter autoclave, and the charged amount of NMP was changed from 6,000 g to 5,000 g, **thereby performing dehydration** over 3.5 hours." (emphasis added).

Clearly, in Example 1a of Miyahara et al, the alkali metal hydrosulfide and the alkali metal hydroxide used in equimolar amounts, in place of the exemplified sodium sulfate, would be 22.49 mol of each and the *additional 30 g of 97% NaOH in the dehydration step* comprises an additional 0.72 mol NaOH, resulting in a NaOH/NaSH ratio of  $(22.49 + 0.72)/22.49$ , which is equal to 1.032. This ratio is outside the NaOH/NaSH range of 0.95 to 1.02 required in the process of claim 5.

Examples 1b, 2 and 5 of Miyahara et al are similar to Example 1a. In Example 3a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 30 g of 97% NaOH (0.72 mol NaOH), resulting in an NaOH/NaSH ratio of  $(21.28 + 0.72)/21.28$ , which is equal to 1.033. Example 3b of Miyahara et al is similar to Example 3a. In Example 4a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 50 g of 97% NaOH (1.21 mol), resulting in an NaOH/NaSH ratio of  $(21.28 + 1.21)/21.28$ , which is equal to 1.056. Example 4b of Miyahara et al is similar to Example 4a. Thus, according to the teachings of Miyahara et al, use of an equimolar amount of sodium hydroxide and sodium hydrosulfide in place of the disclosed sodium sulfide results in an

NaOH/NaSH ratio in the dehydration step above the upper limit of 1.02 required in the process of claim 5.

As an aside, the additional sodium hydroxide which is added in the polymerization step of Miyahara et al, i.e., 7.9 g of 97% NaOH (column 12, line 45) is only used in Applicants' calculation of the sodium hydroxide/charged sulfur source ratio.

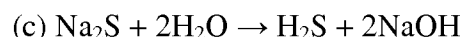
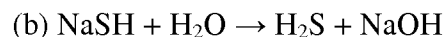
The present specification shows that when the NaOH/NaSH ratio is above 1.02, the properties of the resulting PAS begin to decline. More specifically, in Example 3 in the specification, wherein the NaOH/NaSH ratio is 1.04 before dehydration (comparable to that of Example 1a of Miyahara et al), and the ratio of NaOH to charged sulfur source is 1.075, the PAS yield is lowered, as is the melt viscosity of the PAS, while the yellow index and the amount of dimer impurity are both undesirably increased. Thus, the modification of the Miyahara et al process to include in situ sodium sulfide formation using equimolar amounts of alkali metal hydroxide and alkali metal hydrosulfide does not meet or render obvious the process requirements of claim 5, and the resulting PAS exhibits a less desirable combination of properties.

Applicants note that the Comparative Examples of Miyahara et al do not employ hydrogen sulfide recycle or additional NaOH. However, Miyahara et al do not provide any teaching or suggestion that would provide one of ordinary skill in the art with any reasonable expectation that a proportion of alkali metal hydroxide to alkali metal hydrosulfide in a ratio of 0.95 to 1.02 mol, in combination with the remaining limitations of claim 5, could be used to result in improved properties in the thus formed PAS.

As set forth in detail in Applicants' previous response, if one of ordinary skill in the art were to following the suggestion of Miyahara et al to form sodium sulfide in situ, as the



Examiner has asserted would have been obvious, one of ordinary skill in the art would have naturally considered that at least the following three reactions would occur based on the use of sodium hydrosulfide and sodium hydroxide:



From Miyahara et al, one of ordinary skill in the art would not have known the extent to which each of the above reactions would occur. Further, from Miyahara et al, one of ordinary skill in the art would not have known whether products other than the intended product (sodium sulfide), such as  $\text{H}_2\text{S}$  and  $\text{NaOH}$ , would be contained in the reaction vessel as the result of the step of producing sodium sulfide. In addition, from the Miyahara et al disclosure, one of ordinary skill in the art would not have known what influence these other products would have on subsequent steps, particularly polymerization. One of ordinary skill in the art would normally have feared that these by-products or others in the reaction vessel would adversely affect the PAS product properties. Thus, one of ordinary skill in the art would have considered a step of removing the intended sodium sulfide product from other products obtained in the reaction and dehydration step to isolate sodium sulfide alone for use in the subsequent polymerization.

Presumably, the reason why Miyahara et al did not disclose an example using sodium hydrosulfide and sodium hydroxide “in situ”, despite the knowledge that it would be desirable to use such lower cost reactants, is that various problems have been expected from such a process. This is, in part, described in the present specification as follows:

“There is known a method of using an alkali metal hydrosulfide and an alkali metal hydroxide in combination as materials for the sulfur source. However, this

method is difficult to set conditions for stably performing a polymerization reaction. In addition, according to this method, difficulty is encountered on inhibition of side reactions, the content of volatile matter becomes great, and difficulty is encountered on reduction in the content of bis(4-chlorophenyl) sulfide that is an impurity due to a great amount of the alkali metal hydroxide used upon the polymerization reaction” (page 2, lines 15-25).

Thus, even if one of ordinary skill in the art would have considered producing sodium sulfide “in situ” in the dehydration step, anticipated adverse influences on the subsequent polymerization reaction would have led one of ordinary skill in the art to conduct the in situ formation and dehydration in a different reaction vessel, with separation of the sodium sulfide formed in situ, and Miyahara et al provide no teaching or suggestion that dehydration can be conducted using sodium hydroxide and sodium hydrosulfide as reactants, followed by polymerization in a single reaction vessel as recited in claim 5. Additionally, as noted above, neither Edmunds, Jr. et al nor Campbell resolve these deficiencies as neither teaches a dehydration step using alkali metal hydroxide, alkali metal hydrosulfide and an organic solvent, and, to the contrary, both references show a separate reaction step for alkylaminoalkanoate formation. Thus, there is no expectation that a polymerization reaction as presently claimed can be stably performed using these low cost reactants wherein inconvenient reactions such as thermal decomposition are inhibited (see the present specification at page 6, lines 23-26).

The Examiner asserted in the Official Action that reactions (b) and (c) above which do not produce the desired sodium sulfide require water as a reactant, but the dehydration reaction as taught by Miyahara takes place with the removal of water, thereby diminishing any potential for reactions (b) and (c) to take place, as these reactions require water as a starting material (page 4). Applicants note however that not only is water present during the dehydration step in which these reactions can occur, not all water is removed during the dehydration step and additional water is added at the end of the dehydration step. For example, Miyahara et al disclose at

column 12, lines 34-45, that after dehydration, the reaction vessel contained 22.39 mol  $\text{Na}_2\text{S}$  and 18 g water was added to bring the total water content to 1.04 mol/mol  $\text{Na}_2\text{S}$ . Therefore, the reaction vessel contained 23.29 mol water ( $1.04 \times 22.39$ ) water or 419 g water (18 g/gmole), indicating that 401 g water remained after the dehydration step. Accordingly, the Miyahara et al dehydration system contained water for the above-described reactions (b) and (c).

The Examiner also asserted that the mere fact that a person having ordinary skill in the art may recognize the above reactions as taking place and further assuming that a person having ordinary skill in the art would not know how various side reactions would affect the properties of the PAS produced, does not provide disincentive for preparing the sodium sulfide in situ using sodium hydroxide and sodium hydrosulfide reactants, as Miyahara et al explicitly teach that this is possible (page 4, lines 25-30). However, a person having ordinary skill in the art would have expected a highly alkaline reaction medium due to the use of NaOH and therefore a high likelihood for the reactions (b) and (c) to occur. Additionally, based on the technical common sense at the time the present application was filed, one having ordinary skill in the art would have feared that PAS would not be stably produced due to decomposition of NMP under the highly alkaline conditions and that the quality of PAS would be degraded. Miyahara et al simple provide no technical reasoning to one having ordinary skill in the art at the time the present application was filed to expect that the claimed process not only proceeds stably but also produces a PAS having an advantageous combination of properties.

Importantly, Miyahara et al fail to disclose the claimed ratio of alkali metal hydroxide to alkali metal hydrosulfide of 0.95 to 1.02 mol, the molar ratio of alkali metal hydroxide to charged sulfur source of 1.015 to 1.050, and that hydrogen sulfide formed during the dehydration is discharged to the exterior of the reaction system. While the Examiner may assert that each

difference is minor, these differences in combination with the remaining limitations of claim 5 unexpectedly result in the formation of a complex between alkali metal alkylaminoalkanoate and alkali metal hydrosulfide and allow efficient and economic production of a PAS from reactants consisting essentially of alkali metal hydrosulfide, alkali metal hydroxide, and dihalo-aromatic compound, in organic amide solvent, with a desirable combination of properties. Importantly, Miyahara et al provide no teaching, suggestion or recognition of the formation of a such a complex between the alkali metal alkylaminoalkanoate and the alkali metal hydrosulfide or of significant advantages thereof in the production of PAS in a single reactor.

In determining patentability under 35 U.S.C. §103, it is necessary to determine whether there was an apparent reason to combine known elements in the fashion of the claims at issue, *KSR International Co. v. Teleflex, Inc.*, 550 US 398, 418 (2007). Applicants find no evidence of record which would indicate any apparent reason to one of ordinary skill in the art to modify and supplement the teachings of Miyahara et al to result in the process of claim 5. To the contrary, the formation of an alkylaminoalkanoate and the complex of the alkali metal hydrosulfide therewith according to the process of claim 5, and the stability, efficiency and effectiveness in producing high quality PAS of the claimed process from reactants consisting essentially of alkali metal hydrosulfide, alkali metal hydroxide, and dihalo-aromatic compound, in organic amide, solvent are unexpected and surprising in view of the Miyahara et al teachings, even when viewed in light of Edmunds, Jr. et al and Campbell.

Accordingly, the processes of claim 5, and claims 7, 10-13, and 17-19 dependent thereon, are nonobvious over Miyahara et al and the rejection under 35 U.S.C. §103 has been overcome. Reconsideration is respectfully requested.

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Please charge any fees required in connection with the present communication, or credit any overpayment, to Deposit Account No. 503915.

Respectfully submitted,

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